Contribution ID: **923** Type: **Poster**

Efficient photoreduction strategy for uranium immobilization based on graphite carbon nitride heterojunction nanocomposites

Tuesday, May 17, 2022 5:27 PM (6 minutes)

The conversion of soluble U(VI) to insoluble U(IV) is an economic strategy to efficiently remove U(VI) from radioactive wastewater by photoreduction. However, high photogenerated electron-hole recombination rate and low sunlight utilization are still huge challenges. Here, we designed a carbon nitride-based heterojunction composite material for photocatalytic reduction of U(VI) from aqueous solutions and real seawater systems under simulated sunlight/visible light. The construction mechanism of the heterojunction structure was further clarified through theoretical calculations, and the reduction fixation mechanism was explained with the help of a series of spectral evidence. This strategy of constructing a heterojunction structure effectively inhibits the recombination of photogenerated electrons and holes, and greatly prolongs the lifetime of carriers in the heterojunction system. Therefore, the g-C3N4/LaFeO3 heterojunction exhibits high removal ability in a wide range of U(VI) concentration (460mg/g). Although the construction of g-C3N4/LaFeO3 heterostructure effectively alleviates the problem of rapid recombination of photogenerated electron-hole pairs, the visible light response is still not ideal. Therefore, to enhance the visible light response of the catalyst, we developed a carbon nitride/ceria (CN-CeO2-x) heterojunction with a type II band structure rich in oxygen vacancies based on the heterojunction-defect synergistic modification strategy, used for photoreduction of U(VI) under visible light. CN-CeO2-x heterojunction has excellent photoreduction ability of U(VI) (96.1%), and it can maintain excellent synergistic removal efficiency even in the presence of organic matter that is not conducive to U(VI) removal. The catalytic efficiency of the two catalysts remained at a relatively high level after being used repeatedly for 5 times, indicating that the two photocatalysts have good cost-effectiveness and applicability. In short, g-C3N4/LaFeO3 and CN-CeO2-x heterojunction provide a promising strategy for using inexhaustible solar energy to recover uranium resources.

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Session Classification: Environmental Radioactivity

Track Classification: Radionuclides in the Environment, Radioecology